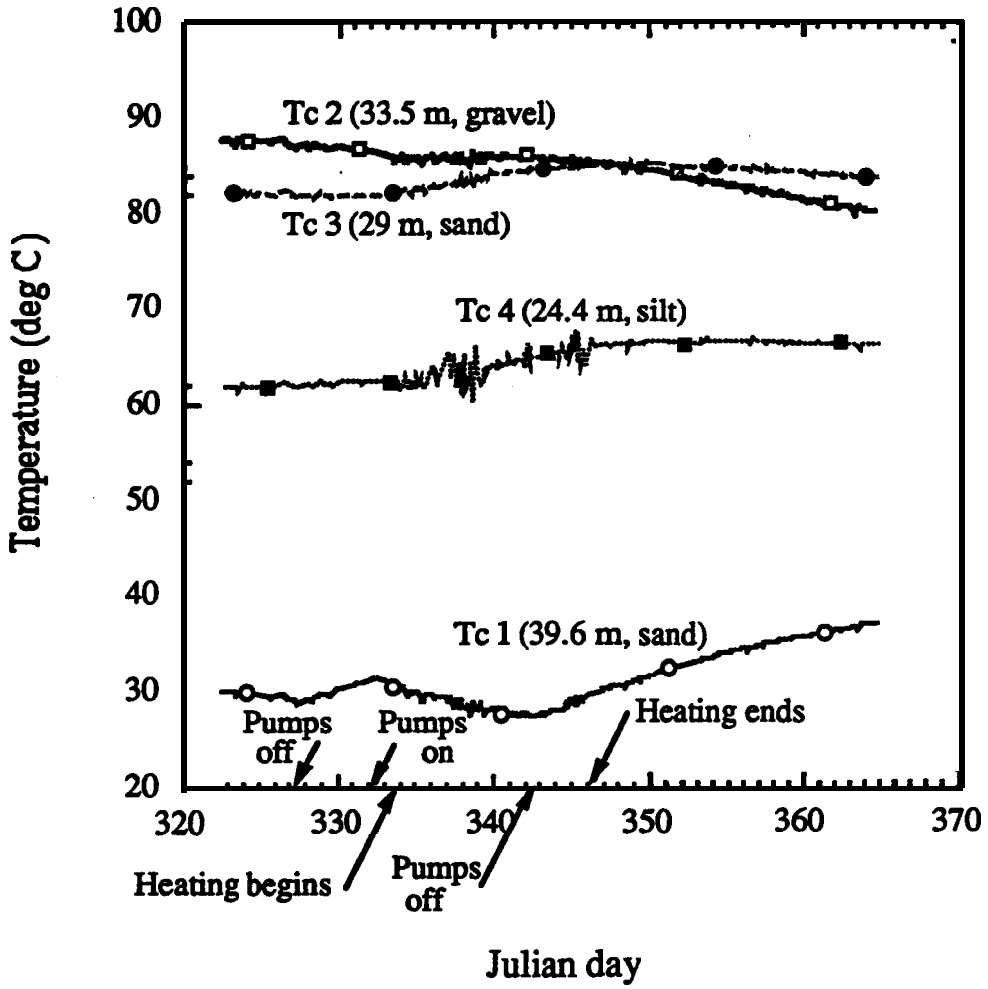


Figure 21. Fixed thermocouples in well TEP 2 show different responses to electrical heating during the ARV phase. Slight temperature rises occur in sandy or silty zones, even while the extraction systems were removing heat from the system. The two lower thermocouples (Tc 1 and 2, open symbols) are located below the standing water table. The two thermocouples above the water table (Tc 3 and 4, closed symbols) show temperature rise throughout electrical heating. (After Sweeney et al., 1994).

Cleanup Results

Free-Product Removal

Free-product gasoline has been removed from the treated area at the LLNL gasoline spill site, thus accomplishing the goal of Dynamic Underground Stripping; approximately 7600 gallons of gasoline were removed from above and below the water table. This conclusion deserves careful scrutiny because of the previous great difficulty in accomplishing this goal experienced by other cleanup methods.

The bases for this conclusion are:

1. 6200 gallons of gasoline were estimated to be in the treatment zone, and 7600 have been removed. After the August 1993 drill-back, soil concentrations indicated that 750 gallons remained (if the distribution was symmetric). Over 1000 gallons have since been removed. Extraction rates fell to nearly zero (11 gallons per month) in January 1994 and have remained low.

2. Groundwater concentrations in the extraction wells and in the two available monitoring wells inside the pattern (GEW 710 and GSW 1A) are lower than the apparent solubility of the most recently extracted gasoline. Although the solubility of gasoline can vary greatly depending on its composition, by measuring the concentration in the water in the oil-water separator where raw

gasoline is known to be in contact with groundwater (Jovanovich et al., 1994; Sweeney et al., 1994), an accurate estimate under site conditions can be obtained. The equilibrium concentrations currently are >35,000 ppb at 20°C; groundwater samples from the extraction and monitoring wells are less than 10,000 ppb at elevated temperatures (>50°C). These are well below the initially observed values for water in contact with free product when wells were drilled (40,000–70,000 ppb), and an order of magnitude below the values observed in the monitoring wells after electrical heating mobilized gasoline (120,000–180,0000 ppb).

3. Vapor and liquid concentrations did not rise significantly after the December 1993–January 1994 shutdown period. Previous shutdowns with hot ground resulted in large increases in concentration when the treatment system was turned on again. Presumably, this was due to the mobilization and/or vaporization of free-product gasoline. The absence of such a pulse after ARV indicates that there was no free product remaining.

4. Groundwater concentrations of BTEX at the central extractors are at lower values than the initial groundwater concentrations just outside the injection ring (e.g., GSW 2, 3, 13), and are at comparable concentrations to many of the distal wells (see below).

The limitations to the conclusion that we have removed all the free product are:

1. Our ability to resolve the presence of free-product pockets by chemical means is limited by the degree of contact with flowing air or water. This is difficult to quantify.

At the start of ARV, the remaining gasoline left a chemical signature of 20,000 ppb total petroleum hydrocarbons (TPH) in groundwater, which dropped to 10,000 ppb by the end of the ARV phase (Sweeney et al., 1994). During ARV, about 1000 gallons of gasoline were removed. This places an upper limit on the free product remaining in the treated area today, based on groundwater analysis alone of 1000 gallons.

There are approximately 1 million gallons of groundwater in the near vicinity of the extraction wells. Given the observed concentrations of TPH during the ARV phase, this places a lower limit of 10-20 gallons (dissolved in groundwater). This indicates that there are much less than 1000, but possibly on the order of tens, of gallons of gasoline remaining (99.9% removal would correspond to about 10 gallons remaining). Any pocket of free product near the extraction wells would have to be extremely well isolated from the permeable parts of the formation to have survived.

2. Free product may remain in the area outside and east of the treated area (e.g., near GSW 216). The vapor concentrations in the eastern-most injection well (GIW 815) are still fairly high (Sweeney et al., 1994). This may be due to either free product in the area or from the vapor being pulled in from the area to the east. It is more likely that this results from vapor being pulled in from the east; if there were free products in the area, we would have seen this in the GSW-001A results. In addition, there was a pocket of free product under the receiving yard to the north of the treatment area before Dynamic Underground Stripping was begun. This was sampled during the drilling of TEP 5 (Bishop et al., 1994).

Groundwater Cleanup

Cleanup of groundwater is the goal of any remediation effort, so the results of the LLNL demonstration must be measured principally in terms of the resulting contaminant concentrations in the water beneath the site even though the goals of the project were strictly limited to free-product removal. The regulated contaminants 1,2 dichloroethane (DCA), xylene, and toluene are at or near their allowed Maximum Contaminant Limit (MCL) in the groundwater of the treated area. Benzene has been reduced dramatically, although it is still well above the MCL (Table 2, Figure 22).

Table 2 gives average values for the major regulated contaminants in the central region of the gas pad; this requires the use of data from several wells, as noted. Dynamic Underground Stripping went far beyond free-product removal; it lowered the benzene concentrations inside the central region to levels below those observed outside the treated area (the so-called bathtub ring of untreated but slightly contaminated water) (Figure 23).

Concentrations of 1,2 DCA have dropped to below detection limits in the treated area, and are significantly reduced in the surrounding region.

Xylene concentrations are diminished in the treated area. The increase in xylene concentration in GSW 216 (east of the treated area) probably reflects the local mobilization of gasoline components through increased solubility and decreased sorption due to heating (Figure 8).

Table 2. Average level of contaminant in central extractors.

Date	Benzene 1.0 ppb		Toluene 100 ppb		Xylenes (1750 ppb)		1,2 DCA (1.0 ppb)		Ethylbenzene (680 ppb)	
	ppb	ratio to mcl -1	ppb	ratio to mcl -1	ppb	ratio to mcl -1	ppb	ratio to mcl -1	ppb	ratio to mcl -1
1987	6400	*6399*	4900	*48.0*	2800	*0.6*	*200*	*399*	*360*	*-0.5*
1988	4600	*4599*	4220	*41.2*	2940	*0.7*	118	*235*	360	*-0.5*
1990	1705	*1704*	1500	*14.0*	1643	*-0.1*	188	*375*	305	*-0.6*
1992 (Pre-DUS)	3646	*3645*	2187	*20.9*	2935	*0.7*	117	*233*	838	*0.2*
1993 (Average DUS)	2061	*2060*	4143	*40.4*	3810	*1.2*	0	*-1*	684	*0.0*
12/93 (Post-DUS)	286	*285*	804	*7.0*	1725	*0.0*	0	*-1*	88	*-0.9*
1/94	170	*169*	683	*5.8*	1866	*0.1*	0	*-1*	36	*-0.9*
3/94	125	*124*	150	*0.5*	846	*-0.5*	0	*-1*	7.7	*-1.0*
6/94	157	*156*	257	*1.6*	327	*-0.8*	0	*-1*	26	*-1.0*
8/94	172	*171*	177	*0.8*	530	*-0.7*	0	*-1*	2	*-1.0*
9/1/1994	209	*208*	189	*0.9*	448	*-0.7*	0	*-1*	6	*-1.0*
Average of wells outside treated area, 1992 *	385	*384*	3	*-1.0*	6	*-1.0*	72	*143*	5	*-1.0*

* Notes: 1987 GSW 15 value from 12/15/87

1988 average of values from GSW-015 in 1988

1990 average of tests of GSW 16 11/6 - 12/14/90

Data for GSW 001A for DCA only, 1990

1992 Average of GEW 816 tests, about 8/15/92

1993 Average of all values observed at SEPI port during second pass operations (from Jovanovich et al., 1994)

12/93 LLNL Lab data sampled 12/6/93 , GO-018. UVI port (although SEPI is consistently about 20% higher)

1/94 Data from LLNL ERD GM-071 sampled 1/19/94, data from UVI port (uncorrected for SEPI/UVI differences if any)

3/94 LLNL lab data GO-091 sampled 3/10/94, UVI port

6/94 LLNL data GP-037 sampled 6/14/94, UVI port. After about 1 month of total shut down.

8/94 LLNL data GP-096 sampled 8/1/94, UVI port.

LLNL data GP-125 sampled 9/1/94, UVI port.

Outside wells: Average of 1992 values for GSW 8,10,208,216 (wells well outside the treated area that had gasoline contaminant)

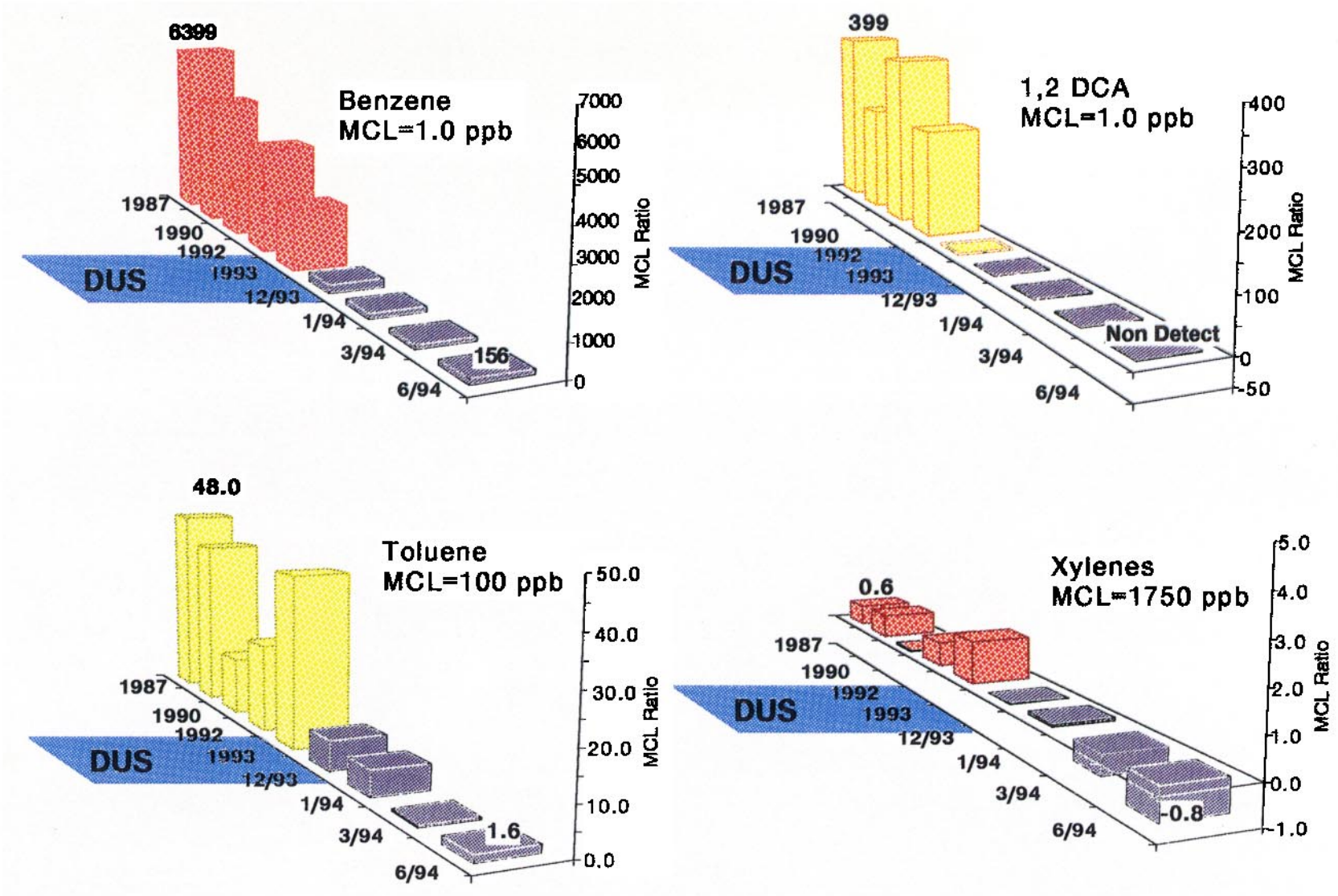


Figure 22. Dissolved groundwater contaminants at the gasoline spill site through June 1994. MCL ratio is expressed as [(contaminant concentration (ppb))/ (MCL (ppb))] – 1. The ratio is zero when the MCL is reached, and drops to negative values (as shown for xylene) when the MCL is exceeded. Values are given for the central extraction wells (GSW 15, 16 and GEW 808, 816). Starting and ending ratios are noted. In June, 1,2 DCA and total xylenes were below MCL, as were ethylbenzene and ethylene dibromide (not shown). Toluene was at 1.6 above its MCL, and benzene was 156 times its MCL. Data from Table 2.

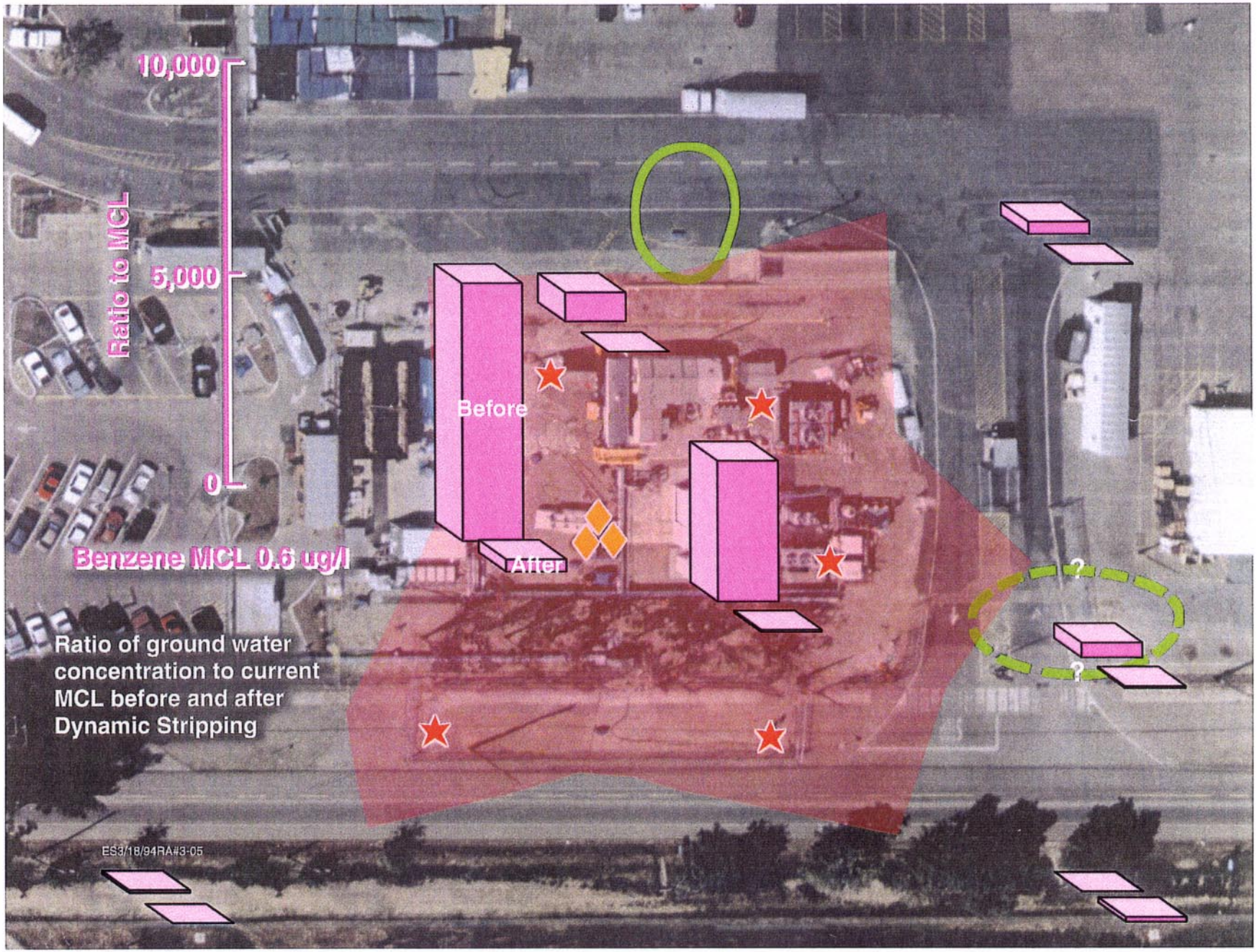
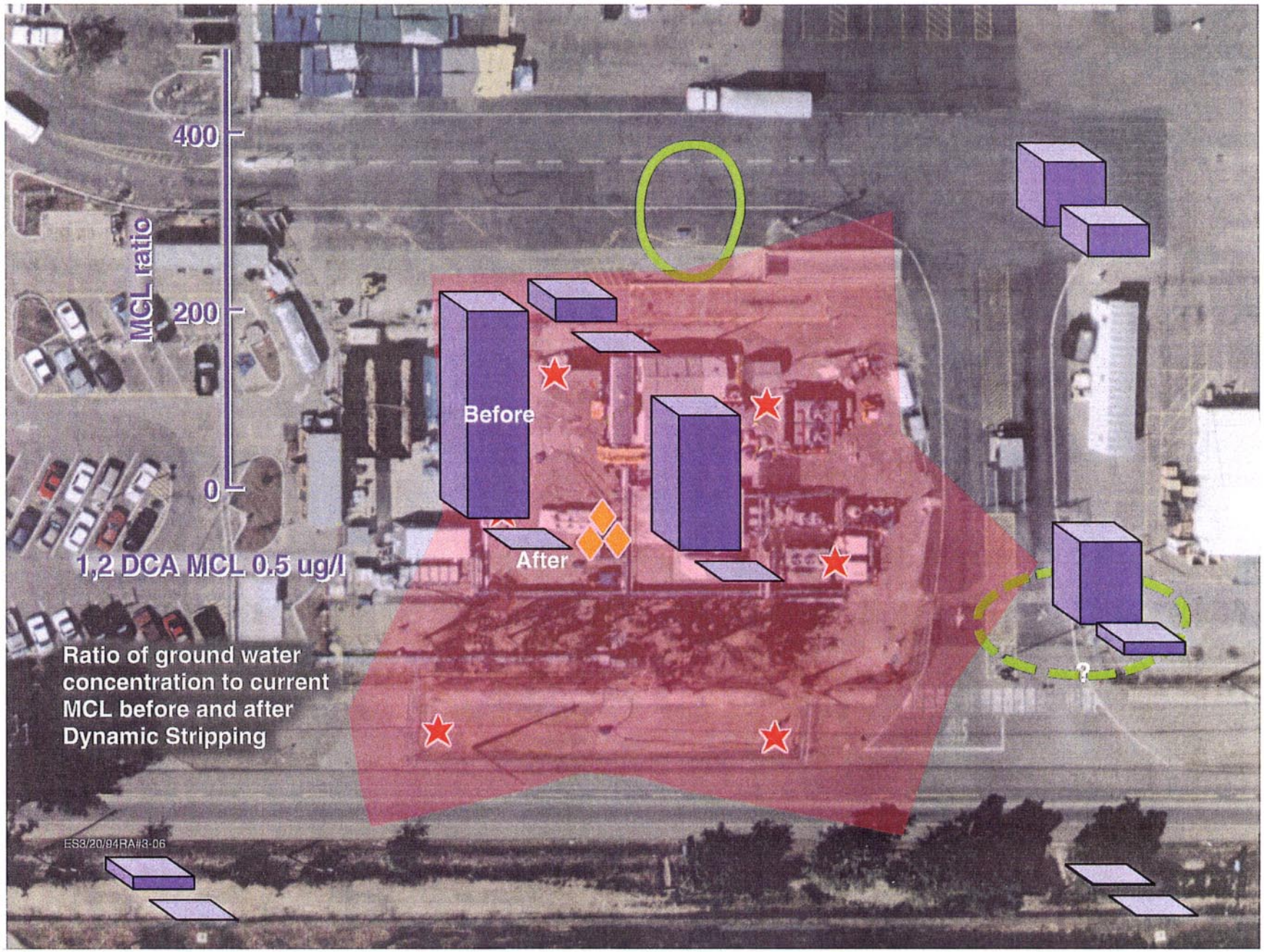
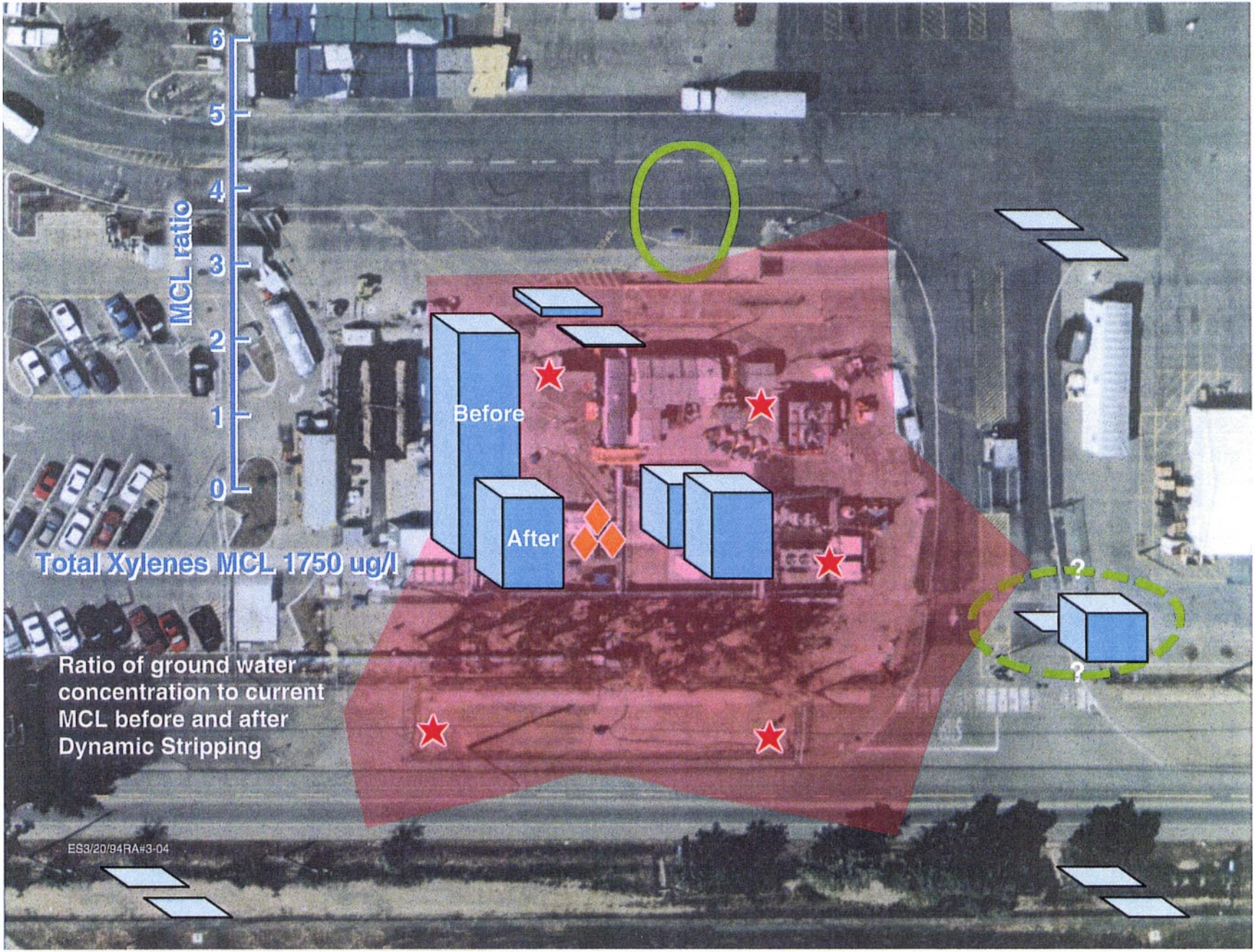


Figure 23. Comparison of MCL ratios observed at seven monitoring or extraction wells before Dynamic Underground Stripping (1992 average values) and after the ARV phase (January 1994). Clockwise from the central extraction wells (GEW 808 and 816) are GEW 710, GSW 208, GSW 216, GSW 7, GSW 8, and GSW 1 (at center of photo) (Figure 6). Free-product gasoline was observed in GSW 216 when it was drilled in 1986 (Dresen et al., 1986). (a) Benzene.


400
MCL ratio
200
0
1,2 DCA MCL 0.5 ug/l
Before
After
Ratio of ground water concentration to current MCL before and after Dynamic Stripping
?
ES3/20/94RA#3-06

(b) 1,2 DCA.



(c) Total xylenes.

Concentrations in GSW 1 appear higher in the treated area; this value roughly matches the levels seen in the extraction wells, and reflects the same mobilization mechanisms. Ethylbenzene and ethylene dibromide are below the MCL as well.

Contaminant concentrations in the central extraction wells appear to approach the outside well values, indicating that water in the treated area is equilibrating with the untreated water as the extraction system draws water in.

The ability of Dynamic Underground Stripping to remove contaminants to such low levels in groundwater is probably indicative of the boil-off distillation mechanism described by Udell (1994a). Because volatile components are generally removed from boiling water at a mass-removal rate exceeding that of the water, boiling of a small percentage of the pore water can dramatically reduce aqueous concentrations.

Udell examines the effect as a function of boiling rate, solubility, and Henry's law constants; unfortunately, solubility and Henry's law constants are not known at high temperatures for most groundwater contaminants (see data for xylene obtained as part of the ARV activities, Sweeney et al., 1994). This mechanism may be responsible for the almost instantaneous removal of 1,2 DCA from the gasoline spill site groundwater by Dynamic Underground Stripping and the dramatic decrease seen in benzene relative to xylene.

Ongoing Bioremediation

Before Dynamic Underground Stripping treatment of the gasoline spill area, a wide variety of microorganisms were actively degrading the BTEX components of the gasoline. These organisms included the dominant genus *Pseudomonas* originally, and after a campaign of vacuum venting in 1990–92, the genus *Flavobactor* was dominant. The largest populations existed in areas where gasoline was present at low concentrations. In the capillary fringe zone (up to 5 ft above the water table) where gasoline concentrations were highest, there were low numbers of culturable organisms. In the central area of the spill, below the water table, oxygen concentrations were very low, and microbial activity was effectively zero.

Extensive characterization of the microbial population was conducted before heating the area, with the expectation that the soils would be sterilized and the population rebound of microorganisms in the area could be studied. Post-test drill-back in August 1993 included collection of extensive soil samples that were cultured for microorganisms both at room temperature and at 50°C.

Although the gram-negative bacteria that had been the dominant BTEX degraders were gone, extensive microbial communities were flourishing in all samples, including those in which the soil was collected at temperatures greater than 90°C. The dominant species were no longer bacteria, but yeasts and related organisms (*Rhodotorula, Streptomyces*), which had been observed in small numbers before heating. Thermophiles previously identified from environments such as the hot springs at Yellowstone National Park are important members of the new community, as well as a number of other organisms apparently representing previously unidentified species.

The rates at which this new biological community are degrading gasoline components has not yet been quantified, but it is clear that BTEX degraders (e.g., *Rhodotorula*) have survived and can rapidly undertake the final removal of contaminants from the groundwater. At this point, the addition of trace nutrients to the system is being considered to enhance this activity. It is hoped that final reduction of benzene levels to below the MCL of 1.0 ppb can be accomplished through a combination of continued intermittent operation of the groundwater and vapor extraction systems to provide oxygen, and proper encouragement of the microbial ecosystem.

Conclusions from the Gasoline Spill Site Demonstration

- Separate-phase gasoline has been removed from the treated area.
- A stable steam zone can be established below the water table.
- Steam injection is effective for heating permeable zones, and repeated steam passes can effectively heat small impermeable layers between.
- Dynamic Underground Stripping can reduce groundwater contamination to very low levels.
- Electrical heating is effective for heating clay zones, but higher power levels are required when extraction of hot fluids is removing heat from the formation.
- Establishing a complete steam zone in very permeable materials requires large amounts of steam; the more the better.
- Electrical resistance tomography is extremely sensitive to heating of soil and gives rapid images of steam movement between wells.
- Tiltmeters accurately mapped the outer extent of the steam fronts both above and below the water table, and the footprint of steam zones emanating from individual injectors in the lower steam zone.
- Steam did not displace much liquid contaminant in a piston flow.
- Vapor recovery is the major contaminant removal mechanism.
- Gasoline is locally mobilized in heated areas and may show higher groundwater concentrations outside the treatment area even though it is not being transported.
- Treatment systems must be robust to handle the large peak extraction rates and the rapid changes in rate.

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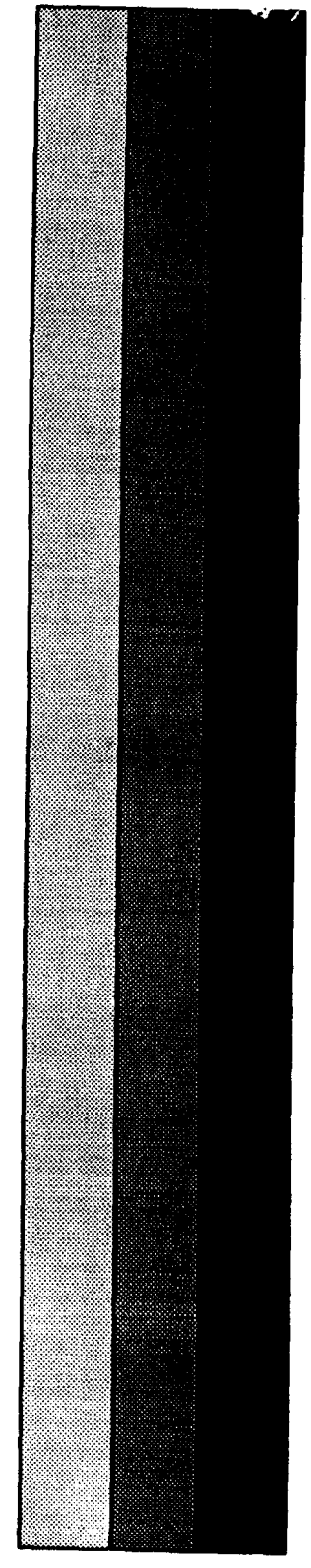

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